

## Effects of acoustic plasmons in photoemission from coupled layered systems

J. D. Lee and A. Fujimori

*Department of Complexity Science and Engineering and Department of Physics, University of Tokyo, Bunkyo-ku, Tokyo 113-0033, Japan*

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We have studied photoemission in a quasi-two-dimensional solid consisting of coupled layers adopting the calculation in the limit of narrow hole band width. Unlike ordinary three-dimensional systems, gapless acoustic plasmons can be excited in the low energy regime. Combining the dielectric response of the coupled layers and the random phase approximation for a single layer, we can explicitly turn on or off the acoustic plasmon excitation. Through the comparison between the on- and off-acoustic calculations, acoustic plasmons in the photoemission spectra are found to suppress the quasiparticle weight and lead to an asymmetric broadening of the quasiparticle peak. Such tendencies are strengthened as the interlayer dielectric coupling increases. Further, the model is applied to the high-temperature superconductors and found to give additional asymmetric broadening to the pure two-dimensional spectra, which means that a significant part of the anomalously large inelastic background may be attributed to multiple intrinsic losses by acoustic plasmons.

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### I. INTRODUCTION

Angle-resolved photoemission spectroscopy (ARPES) has contributed significantly to the understanding of correlated systems by delivering valuable informations about their electronic structures. Particularly, due to the inherent momentum conservation in a crystal, ARPES has been a more powerful tool to study correlated two-dimensional systems like high-temperature (high- $T_C$ ) superconductors.<sup>1</sup> For such systems, theoretical studies have been concentrated on the purely two-dimensional correlated model and proposed Fermi-liquid (FL), marginal Fermi-liquid (MFL),<sup>2</sup> or Luttinger liquid<sup>3</sup> behaviors in interpreting the experimental data. However, it should be noted that the studies have *a priori* neglected the three dimensionality of the actual system due to dielectric coupling between the layers, even if electron transfer is negligible between the layers and many physical properties can be explained by considering the two-dimensional  $\text{CuO}_2$  layer.

One of the most distinct features in the strongly correlated system that cannot be understood within the weak correlation picture is the absence or suppression of the quasiparticle component in ARPES.<sup>1</sup> Going to the stack of interacting layers beyond the purely two-dimensional model, we meet unusual gapless acoustic plasmons not shown in ordinary three-dimensional systems.<sup>4</sup> The *gapless* property is very interesting in photoemission because it can produce multiple low energy losses. Along this line, the gapless magnons have been studied for one- or two-dimensional undoped Mott-Hubbard insulators and found to highly suppress the coherent component (corresponding to the quasiparticle component) of the single-particle spectral function.<sup>5</sup> In many cases, however, the doped cuprates exhibit the opening of spin gap, which means magnons may be no longer gapless in the interesting doped cases.<sup>6</sup> In such a sense, the acoustic plasmon is more interesting because the gaplessness of acoustic plasmons comes from the dielectric coupling among the layers and should be robust with respect to the doping as long as each layer has mobile electrons. In addition to the high- $T_C$  cuprates, excitations due to coupling among layers, i.e., three

dimensionality of the system, may be potentially important in another layered compounds, for instance, transition-metal dichalcogenides  $\text{TiSe}_2$ ,  $\text{TiS}_2$ , or  $\text{TiTe}_2$ .<sup>7</sup>

Recently, Hedin and Lee suggested the model of the stack of two-dimensional layers and studied the photoemission in the model.<sup>8</sup> They have used the experimental energy loss spectra for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  (Bi2212) (Ref. 9) as the many-body input. In their approach, however, the relevant excitations are so completely melted into the experimental data that the role of acoustic plasmons or their dispersing properties cannot be explicitly specified or scrutinized even if one can expect *some* effects from acoustic plasmons.

In this paper, we combine the dielectric response of coupled layers and the RPA expression for a layer. Then one can explicitly turn on or off the acoustic plasmon excitation, which is very useful in exploring the effects of acoustic plasmons because one can directly compare the on- and off-acoustic cases. Further, to simulate the realistic situation, we introduce two dielectric constants  $\epsilon_{\parallel}$  and  $\epsilon_{\perp}$  for the screened Coulomb potential.<sup>10</sup> It has then been found that acoustic plasmons suppress the quasiparticle peak weight and increase broadening more asymmetrically similar to the case of gapless magnons.<sup>5</sup> It is better understood by fitting the quasiparticle components with the Doniach-Sunjc line shape<sup>11</sup> characterized by the singularity index. As the interlayer coupling increases, acoustic plasmons are more strongly activated in the photoemission. There have been various theoretical studies about the estimation of the quasiparticle component in the system,<sup>12</sup> however, the controversies as to whether such theories can explain the experimentally observed dramatic reduction of the quasiparticle weight are still going on. It should be pointed out that acoustic plasmons decrease the quasiparticle weight beyond the two-dimensional model. Further, the present model can be naturally applied to the high- $T_C$  superconductors and is found to give additional significant asymmetric broadening to the pure two-dimensional spectra described by the MFL (or FL) theory. This may explain the anomalously large inelastic background needed to fit the experimental data with the MFL or FL theory.<sup>13</sup>

The paper is organized as follows. In Sec. II, the basic formalism is briefly described. In Sec. III, the discussion on the role of acoustic plasmons comes through comparison between the on- and off-acoustic calculations. In Sec. IV, we discuss an application of the present model to the high- $T_C$  superconductors and its significance. Concluding remarks are given in Sec. V.

## II. BASIC FORMALISM

In the strongly correlated system consisting of coupled layers, the final photoemission spectrum  $J_{\mathbf{k}}(\omega)$  is found to be<sup>8</sup>

$$J_{\mathbf{k}}(\omega) = \int d\omega' J_{\mathbf{k}}^{2D}(\omega') P(\omega - \omega'), \quad (1)$$

where  $J_{\mathbf{k}}^{2D}(\omega')$  is the spectral function of the pure two-dimensional layer and  $P(\omega)$  is the effect of the coupling among layers, that is,  $P(\omega)$  is regarded as being from the three dimensionality of the system.<sup>14</sup> If one puts  $J_{\mathbf{k}}^{2D}(\omega) = \delta(\omega - \varepsilon_{\mathbf{k}})$ ,  $P(\omega)$  is just the final photoemission spectrum and otherwise  $P(\omega)$  may play a role of the loss function. In this study, we are mainly interested in  $P(\omega)$  where the acoustic plasmon enters. Under the quasiboson model<sup>15</sup> and the narrow hole band width limit,  $P(\omega)$  is determined from the dielectric response of the system. The narrow hole band width means that the calculation would follow the core hole case even if we have in our mind the valence band photoemission, that is, recoil effects of the valence hole would be neglected. Its practical justifications are found in Refs. 16,17, and 18.

In the quasiboson model,  $P(\omega)$  is given by

$$P(\omega) = \frac{1}{2\pi} \int dt e^{i\omega t} \exp \left[ \int d\omega' \alpha(\omega') \frac{e^{i\omega' t} - 1}{\omega'} \right] \quad (2)$$

and  $\alpha(\omega)$  is

$$\alpha(\omega) = \frac{-\omega}{\pi A} \sum_{\mathbf{Q}} \int dz dz' f(z) f(z')^* \text{Im} W(z, z', \mathbf{Q}; \omega), \quad (3)$$

where  $f(z) = -\delta(z - z_0)/\omega$  ( $z_0$ : hole position) within the sudden approximation and  $\mathbf{Q}$  is the two-dimensional vector parallel to the layer and  $z$  or  $z'$  is the coordinate perpendicular to the layer ( $z > 0$  is occupied by the solid).  $P(\omega)$  is obtained by a summation of the limited diagram set up to the infinite order and exact up to the second order of the fluctuation potential. In addition,  $P(\omega)$  is known to properly describe the quasiparticle parts ( $\omega \approx 0$ ).<sup>15</sup>  $W(z, z', \mathbf{Q}; \omega)$  in Eq. (3) is the dynamically screened potential and its Fourier transform for  $\mathbf{q} = (q, \mathbf{Q})$  satisfies

$$W(q, q', \mathbf{Q}; \omega) = v(q, \mathbf{Q}) \epsilon^{-1}(q, q', \mathbf{Q}; \omega). \quad (4)$$

It is then clear that the dielectric response of the system would determine the photoemission spectra of the localized hole.

For simplicity, we think of  $\text{Bi}_2\text{Sr}_2\text{CuO}_6$  (Bi2201) having one  $\text{CuO}_2$  layer per unit cell as a reference material. If  $c$  is

the lattice constant (or the interlayer distance) along the  $c$  axis, the inverse dielectric function of the system is

$$\begin{aligned} \epsilon^{-1}(q, q', \mathbf{Q}; \omega) \\ = 1 - v(\mathbf{q}) \frac{\chi^0(\mathbf{Q}; \omega)/c}{1 - V(q, \mathbf{Q}) \chi^0(\mathbf{Q}; \omega)/c} \sum_G \delta_{q+G, q'}, \end{aligned} \quad (5)$$

where  $G = 2n\pi/c$  is the reciprocal lattice vector along the  $c$  axis,  $v(\mathbf{q})$  is the screened Coulomb potential given by

$$v(\mathbf{q}) = 4\pi \frac{1}{q^2 \epsilon_{\perp} + \mathbf{Q}^2 \epsilon_{\parallel}}, \quad (6)$$

and  $V(q, \mathbf{Q})$  is

$$V(q, \mathbf{Q}) = \sum_G v(q + G, \mathbf{Q}) = \frac{2\pi c}{\zeta Q} \frac{\sinh \xi Q c}{\cosh \xi Q c - \cos qc}. \quad (7)$$

In Eqs. (6) and (7), we define  $\xi \equiv (\epsilon_{\parallel}/\epsilon_{\perp})^{1/2}$  and  $\zeta \equiv (\epsilon_{\parallel}\epsilon_{\perp})^{1/2}$ . We remark that  $\chi^0(\mathbf{Q}; \omega)$  is the polarization function for a noninteracting layer. The RPA expression will be adopted for  $\chi^0(\mathbf{Q}; \omega)$  in the approach. Using Eqs. (4) and (5) and taking the Fourier transformation of  $W(q, q', \mathbf{Q}; \omega)$ , Im  $W(z, z', \mathbf{Q}; \omega)$  is given by

$$\begin{aligned} \text{Im} W(z, z', \mathbf{Q}; \omega) &= \frac{1}{2\pi} \int_0^{\pi/c} dq V_f^r(q, \mathbf{Q}, z) V_f^r(q, \mathbf{Q}, z') \\ &\times \text{Im} \chi_{\text{scr}}(q, \mathbf{Q}; \omega), \end{aligned} \quad (8)$$

$$\chi_{\text{scr}}(q, \mathbf{Q}; \omega) = \frac{\chi^0(\mathbf{Q}; \omega)/c}{1 - V(q, \mathbf{Q}) \chi^0(\mathbf{Q}; \omega)/c}. \quad (9)$$

The fluctuation potential  $V_f^r(q, \mathbf{Q}, z)$  is defined by adding the phase factor  $e^{i\phi}$  to  $V_f(q, \mathbf{Q}, z)$ ,

$$V_f^r(q, \mathbf{Q}, z) = 2 \text{Re}[V_f(q, \mathbf{Q}, z) e^{i\phi}],$$

$$\begin{aligned} V_f(q, \mathbf{Q}, z) &= \frac{2\pi c}{\zeta Q} \frac{\sinh \xi Q c (1 - \delta) + e^{-iqc} \sinh \xi Q c \delta}{\cosh \xi Q c - \cos qc} \\ &\times e^{-iq(z - c/2 - c\delta)}, \end{aligned} \quad (10)$$

where  $\delta$  is defined as  $\text{res}(z - c/2, c)/c$  and the charged layer is assumed to be at  $z = (n + 1/2)c$  ( $n$  is an integer,  $\geq 0$ ), consistent with the structure of Bi2201. The phase factor  $e^{i\phi}$  should be determined to satisfy the surface boundary condition,  $V_f^r(q, \mathbf{Q}, 0) = 0$  (when  $z = 0$  is put the surface),

$$\cos(qc + \phi) + \cos \phi = 0, \quad \text{i.e. } \phi = \frac{\pi}{2} - \frac{qc}{2}, \quad (11)$$

where we used  $\delta = 1/2$  at  $z = 0$ . More details on the fluctuation potential are found in Ref. 8 for the similar system.

### III. ACOUSTIC PLASMONS IN PHOTOEMISSION

In this section we concentrate on the study of  $P(\omega)$ . As mentioned in the last section,  $P(\omega)$  may be considered as the true photoemission spectra when  $J_{\mathbf{k}}^{2D}(\omega)$  is assumed as a sum of pure delta functions.

We note that, putting  $z = Q/2k_F$  and  $u = \omega/Qk_F$ ,  $\chi^0(\mathbf{Q}, \omega)$  within RPA is given by<sup>19</sup>

$$\text{Re } \chi^0(\mathbf{Q}, \omega) = G \{ 2z - C_- [(z-u)^2 - 1]^{1/2} - C_+ [(z+u)^2 - 1]^{1/2} \},$$

$$\text{Im } \chi^0(\mathbf{Q}, \omega) = G \{ D_- [1 - (z-u)^2]^{1/2} - D_+ [1 - (z+u)^2]^{1/2} \}, \quad (12)$$

where  $G = -k_F/Q\pi$ ,  $C_{\pm} = (z \pm u)/|z \pm u|$  and  $D_{\pm} = 0$  for  $|z \pm u| > 1$ , and  $C_{\pm} = 0$  and  $D_{\pm} = 1$  for  $|z \pm u| < 1$ .

From Eq. (12), when  $\omega \gg |\mathbf{Q}|k_F$  ( $s^2 = k_F^2/2$ ),  $\chi^0(\mathbf{Q}, \omega)$  becomes

$$\chi^0(\mathbf{Q}; \omega) = n \frac{\mathbf{Q}^2}{\omega^2 - s^2 \mathbf{Q}^2},$$

where  $n$  is the electron density of a layer. Collective excitations of the system are found from the pole structure of  $\text{Im } \epsilon^{-1}(q, \mathbf{Q}; \omega)$ .<sup>20</sup> As the pole is determined by the zero of  $1 - V(q, \mathbf{Q})\chi^0(\mathbf{Q}; \omega)/c$ , we get

$$\omega^2 = s^2 \mathbf{Q}^2 + \frac{2n\pi}{\zeta} Q \frac{\sinh \xi Q c}{\cosh \xi Q c - \cos qc}, \quad (13)$$

where we find, when  $\mathbf{Q}$  is small, (i)  $q=0$ ,  $\omega^2(\mathbf{Q}) \approx s^2 \mathbf{Q}^2 + 4\pi n/\epsilon_{\parallel} c$  (optical plasmon) and (ii)  $q \neq 0$ ,  $\omega^2(\mathbf{Q}) \approx s^2 \mathbf{Q}^2$

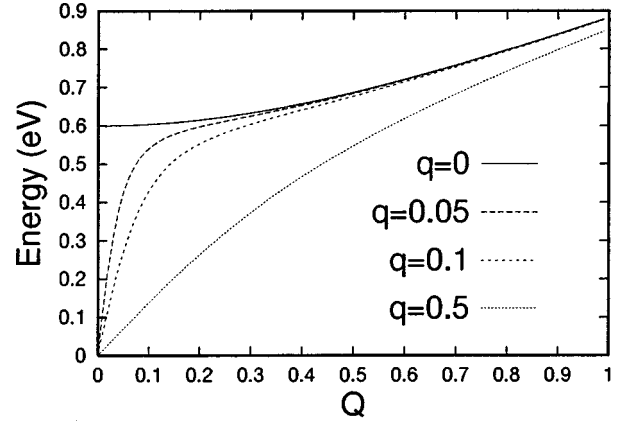


FIG. 1. Depending on  $q$ , plasmon dispersions are given with respect to  $|\mathbf{Q}|$ . The unit of  $|\mathbf{Q}|$  or  $q$  is  $\pi/c$ .

(acoustic plasmon). In Fig. 1, plasmon energy dispersions are given for a few values of  $q$ 's. For the evaluation, the optical plasmon frequency  $4\pi n/\epsilon_{\parallel} c = 0.6$  eV,  $\epsilon_{\parallel} = \epsilon_{\perp} = 4$ , and  $c = 12.18$  Å (Ref. 21) for Bi2201 are used.

Substituting Eqs. (8) and (9) for Eq. (3),  $\alpha(\omega)$  is recast into

$$\alpha(\omega) = - \frac{1}{\pi(2\pi)^3} \frac{1}{\omega} \int_0^{\pi/c} dq \int d\mathbf{Q} [V_f^r(q, \mathbf{Q}; z_0)]^2 \times \text{Im}[\chi_{\text{scr}}(q, \mathbf{Q}; \omega)], \quad (14)$$

and  $\text{Im } \chi_{\text{scr}}(q, \mathbf{Q}; \omega)$  governs the energy loss by making excitations in the stack of layers

$$\text{Im } \chi_{\text{scr}}(q, \mathbf{Q}; \omega) = \frac{\text{Im } \chi^0(\mathbf{Q}, \omega)/c}{[1 - V(q, \mathbf{Q})\text{Re } \chi^0(\mathbf{Q}, \omega)/c]^2 + [V(q, \mathbf{Q})\text{Im } \chi^0(\mathbf{Q}, \omega)/c]^2}.$$

It is interesting and important to note that, by putting  $q=0$  in  $\text{Im } \chi_{\text{scr}}(q, \mathbf{Q}; \omega)$ , one can turn off the excitation of acoustic plasmons. Here we define the off-acoustic plasmon calculation for  $\tilde{\alpha}(\omega)$  as

$$\tilde{\alpha}(\omega) = - \frac{1}{\pi(2\pi)^3} \frac{1}{\omega} \int_0^{\pi/c} dq \int d\mathbf{Q} [V_f^r(q, \mathbf{Q}; z_0)]^2 \times \text{Im}[\chi_{\text{scr}}(q=0, \mathbf{Q}; \omega)]. \quad (15)$$

In Fig. 2, typical behaviors of  $\alpha(\omega)$  and  $\tilde{\alpha}(\omega)$  are given. Qualitatively,  $\tilde{\alpha}(\omega)$  is similar to that of the ordinary three-dimensional electron gas without acoustic plasmons,<sup>15</sup> which signifies that in  $\tilde{\alpha}(\omega)$  acoustic plasmons are pertinently deleted. From  $\alpha(\omega)$  [or  $\tilde{\alpha}(\omega)$ ] we can readily evaluate  $P(\omega)$ , for which we always consider only the case leaving a hole in

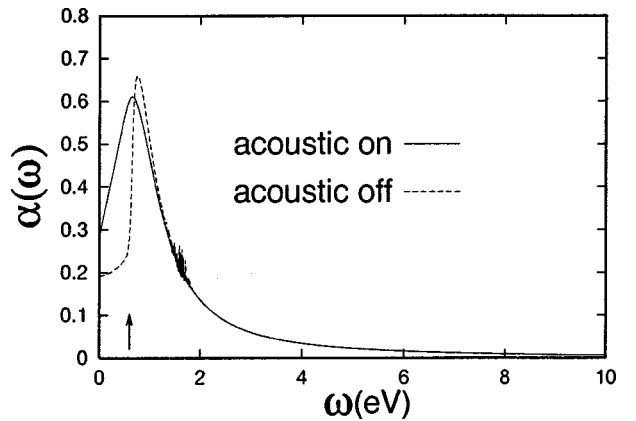


FIG. 2. Typical behaviors of  $\alpha(\omega)$  (acoustic on) and  $\tilde{\alpha}(\omega)$  (acoustic off) are given for  $\epsilon_{\parallel} = \epsilon_{\perp} = 4$ . The arrow indicates the energy of  $4\pi n/\epsilon_{\parallel} c$ .

the first charged ( $\text{CuO}_2$ ) layer, i.e.,  $z_0 = c/2$  [ $\mathbf{r}_0 = (z_0, \mathbf{0})$ ]. Then we note that  $V_f^r(q, \mathbf{Q}; z_0)$  is

$$V_f^r(q, \mathbf{Q}; z_0) = \frac{4\pi c}{\zeta Q} \frac{\sinh \xi Q c}{\cosh \xi Q c - \cos qc}.$$

$V_f^r(q, \mathbf{Q}; z_0)$  corresponds to the core-hole potential in the core-level photoemission problem and plays a role of the driving force for the nontrivial losses in the photoemission. It is easily seen that, when  $V_f^r(q, \mathbf{Q}; z_0)$  vanishes,  $P(\omega)$  reduces to the trivial  $\delta(\omega)$ . It is also worth noting that  $P(\omega)$  is determined solely by values of  $\alpha(\omega')$  or  $\tilde{\alpha}(\omega')$  at  $\omega' \leq \omega$  and then the quasiparticle line shape for the off-acoustic case is determined only by the particle-hole contribution.

The present study is based on the nonrecoiling valence hole by assuming the small hole band width, similar to the core-level case. It is well known that the core-level PES from metals should have an asymmetric skew line shape due to low energy excitations, that is, the Doniach-Sunjc line shape  $A_{\text{DS}}(\omega)$ .<sup>11</sup> Through detailed studies allowing the recoiling of the valence hole in metals, valence hole spectra are found so asymmetric as to be comparable to core hole spectra.<sup>17</sup> Further, this analysis has been successfully applied to the valence band spectra of Ti, Zr, Nb, Mo, and Hf by Hüfner and his co-workers.<sup>18</sup> Therefore, it is expected that the quasiparticle part of  $P(\omega)$  would be reasonably fitted with Doniach-Sunjc line shape. The  $\Gamma$ -broadened Doniach-Sunjc spectra  $A_{\text{DS}}(\omega)$  is

$$A_{\text{DS}}(\omega) \propto \frac{\cos[\pi\alpha_0/2 + (1-\alpha_0)\tan^{-1}(\omega/\Gamma)]}{[1 + (\omega/\Gamma)^2]^{(1-\alpha_0)/2}}, \quad (16)$$

where  $\alpha_0$  is the singularity index. In Fig. 3, the spectral functions of  $P(\omega)$  and their corresponding fitted Doniach-Sunjc spectra are given for on- and off-acoustic cases, respectively. The on-acoustic case is just a full calculation. We find that the fitting is very nice especially for the quasiparticle parts. The necessary  $\alpha_0$ 's are found to be 0.202 and 0.119 for on- and off-acoustic calculations. The latter 0.119 is attributed to the electron-hole excitation. The interlayer coupling strength is controlled by  $\epsilon_{\perp}$ . As  $\epsilon_{\perp}$  decreases, the interlayer coupling increases, and vice versa, which can be easily understood by expressing Eq. (6) in the real-space coordinates as

$$V(z, \mathbf{R}) = \frac{1}{(\epsilon_{\parallel}^2 z^2 + \epsilon_{\parallel} \epsilon_{\perp} \mathbf{R}^2)^{1/2}},$$

where one sees that, as  $\epsilon_{\perp} \rightarrow 0$ ,  $V(z, \mathbf{R}) \rightarrow 1/\epsilon_{\parallel} z$ , on the other hand, as  $\epsilon_{\perp} \rightarrow \infty$ ,  $V(z, \mathbf{R}) \rightarrow 1/\zeta |\mathbf{R}|$ . This is also reflected in the fluctuation potential  $V_f^r(q, \mathbf{Q}; z_0)$ . In the limit of  $\epsilon_{\perp} \rightarrow \infty$  (extinguishing the interlayer coupling),  $V_f^r(q, \mathbf{Q}; z_0)$  survives for only  $q=0$  mode that is for the optical plasmon. This is consistent since one can expect only the optical plasmon mode in the case. We always fix  $\epsilon_{\parallel}$  through  $4\pi n/\epsilon_{\parallel} c = 0.6$  eV in the following discussion because  $4\pi n/\epsilon_{\parallel} c$  is a meaningful energy scale of the system.

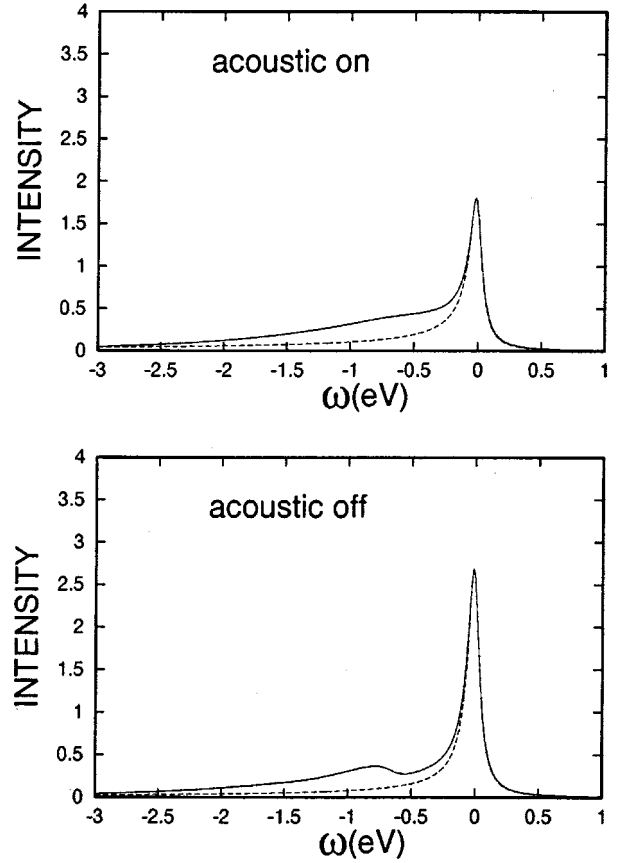


FIG. 3.  $P(\omega)$  calculated with acoustic plasmons on and off. The on-acoustic calculation (upper panel) is  $P(\omega)$  from the full expression of Eq. (14), while the off-acoustic calculation (lower panel) from  $\tilde{\alpha}(\omega)$  of Eq. (15) for the typical  $\epsilon_{\parallel} = \epsilon_{\perp} = 4.0$ . The dashed lines are fit by the Doniach-Sunjc  $A_{\text{DS}}(\omega)$ . The broadening of  $\Gamma = 50$  meV is applied.

It is interesting to investigate the behaviors of  $P(\omega)$  with respect to the interlayer coupling strength, i.e.,  $\epsilon_{\perp}$  for a fixed  $\epsilon_{\parallel}$ . In Fig. 4, we provide the behaviors of  $P(\omega)$  as  $\epsilon_{\perp}$  varies from 1.0 to 5.0. It is noted that there are drastic behaviors for both on- and off-acoustic cases for a given range of  $\epsilon_{\perp}$ . Particularly, for the on-acoustic case with a small  $\epsilon_{\perp}$ , say  $\epsilon_{\perp} = 1$ , the quasiparticle part is highly suppressed. It is very useful to estimate the quasiparticle peak weight from the fitting through the Doniach-Sunjc spectral line shape even if the estimation may be less accurate as  $\epsilon_{\perp}$  (or  $\alpha_0$ ) becomes smaller. For such estimations, we need to set the cutoff energy for  $A_{\text{DS}}(\omega)$  because its integration is not well defined when  $\omega \rightarrow -\infty$ . Here we take the cutoff energy as  $\approx -0.6$  eV, the characteristic energy where the optical plasmon begins to excite.

Figure 5 gives the systematic profile of the quasiparticle peak weight and singularity index with  $\epsilon_{\perp}$  varied from 1.0 to 5.0. The larger singularity index means the more asymmetric broadening. Taking the ratio of quasiparticle weight of on-acoustic calculations to off-acoustic ones, we find the ratio decreases from  $\sim 0.836$  to  $\sim 0.727$  as the interlayer coupling increases (from  $\epsilon_{\perp} = 5.0$  to 1.0). This directly shows that as the interlayer coupling strengthens, the acoustic plasmons

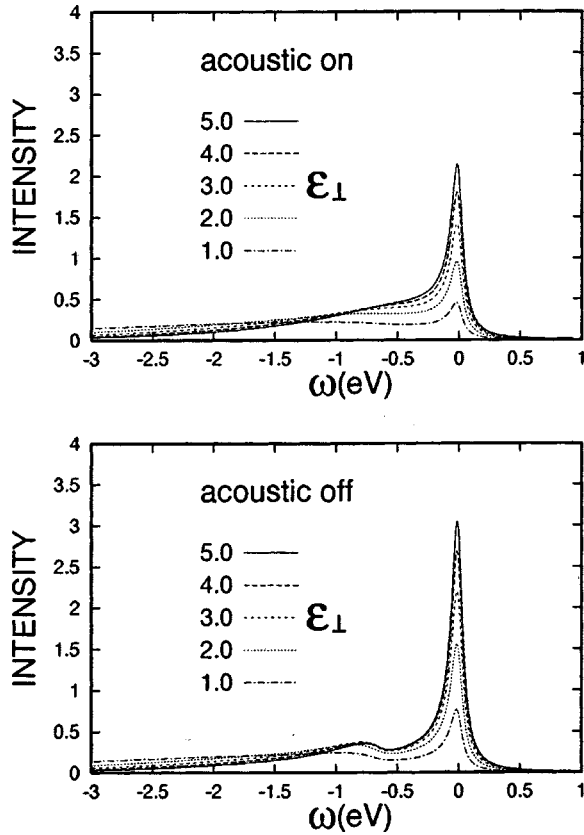


FIG. 4. Behaviors of  $P(\omega)$  with respect to  $\epsilon_{\perp} = 1.0-5.0$  for a fixed  $\epsilon_{\parallel} = 4.0$  (also fixing  $4\pi n/\epsilon_{\parallel}c = 0.6$  eV). The 50 meV broadening is applied.

are more vigorously activated in the photoemission process. On the other hand, in the opposite limit, i.e.  $\epsilon_{\perp} \rightarrow \infty$ , only the optical plasmon activates and then the ratio goes to 1.0.

#### IV. HIGH- $T_C$ SUPERCONDUCTORS

Although the RPA approach for the layered conducting system immediately gives the acoustic plasmons as described in the last section, it is true that no direct experimental observation of acoustic plasmons in the high- $T_C$  superconduct-

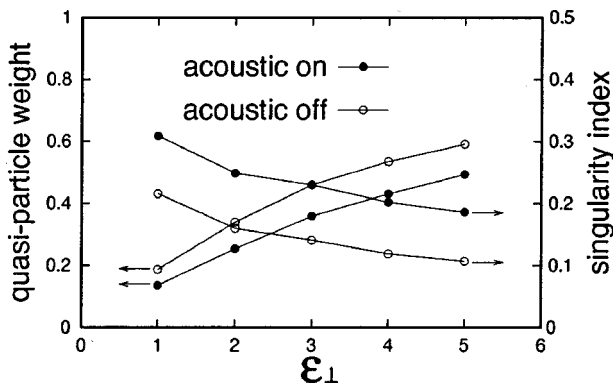


FIG. 5. Quasiparticle weight and singularity indexes with respect to  $\epsilon_{\perp}$  for both on- and off-acoustic calculations.  $\epsilon_{\parallel} = 4$  is fixed.

ors has been reported so far. Optical measurements<sup>22</sup> and the transmission electron energy loss spectroscopy (EELS) experiments<sup>9</sup> fail in directly observing them. The reason seems to be in that those experiments are done for  $\mathbf{q} \approx 0$  or  $q \approx 0$ , where the energies of acoustic plasmons are very small. However, recent reflection EELS experiments deliver the strong possibility to establish their existence since it allows a measurement as a function of  $\mathbf{Q}$ , while summing all perpendicular momenta  $q$ .<sup>23</sup>

In high- $T_C$  superconductors,  $J_{\mathbf{k}}^{2D}(\omega)$  is not a simple  $\delta(\omega - \epsilon_{\mathbf{k}})$  any longer, but is a spectral function with the characteristic self-energy. This is a difference from the discussion in the last section. In many cases, the ARPES data of high- $T_C$  compounds are fitted using the marginal Fermi liquid (MFL) theory.<sup>2</sup> In the theory, the spectral function is  $[ = -(1/\pi)\text{Im} G(\mathbf{k}, \omega)$

$$J_{\mathbf{k}}^{2D}(\omega) = \frac{1}{\pi} \frac{-\text{Im} \Sigma(\mathbf{k}, \omega)}{[\omega - \epsilon_{\mathbf{k}} - \text{Re} \Sigma(\mathbf{k}, \omega)]^2 + [\text{Im} \Sigma(\mathbf{k}, \omega)]^2}, \quad (17)$$

where the self-energy  $\Sigma(\mathbf{k}, \omega)$  is, when neglecting the  $\mathbf{k}$  dependency,

$$\Sigma(\mathbf{k}, \omega) \sim g'^2 \left( \omega \ln \frac{x}{\omega_c} - i \frac{\pi}{2} x \right), \quad x = \max(|\omega|, T). \quad (18)$$

On the other hand, the self-energy of the Fermi-liquid (FL) theory has the form of  $\Sigma(\mathbf{k}, \omega) \sim -\alpha\omega - i\beta\omega^2$  near  $\omega \sim 0$ .

Liu, Anderson, and Allen<sup>13</sup> have tried to fit the ARPES data of high- $T_C$  compounds using both MFL and FL theories and concluded that neither MFL nor FL theories can fit the slow fall-off (broadness) of the spectra unless the anomalously large inelastic backgrounds are assumed.

It is very interesting to note that  $P(\omega)$  in the last section can give another effects on  $J_{\mathbf{k}}(\omega)$  from Eq. (1). Although Liu *et al.* tried to fit the MFL or FL to Bi2212 having two  $\text{CuO}_2$  layers per unit cell and our calculation of  $P(\omega)$  is basically for Bi2201 having a single layer, the application of  $P(\omega)$  is thought still meaningful at least from a qualitative point of view. The necessary on-acoustic (full)  $P(\omega)$  is evaluated from  $\epsilon_{\parallel} = \epsilon_{\perp} = 5.0$ , which are actually reasonable values for transition-metal oxides including high- $T_C$  materials.<sup>24</sup>

In Fig. 6, we give a few curves for  $\epsilon_{\mathbf{k}} = -0.1, -0.3, -0.5,$  and  $-0.7$  eV, where the actual quasi-particle energies are determined by  $\omega - \epsilon_{\mathbf{k}} - \text{Re} \Sigma(\mathbf{k}, \omega) = 0$  as  $-0.020, -0.087, -0.187,$  and  $-0.334$  eV. Because we neglect the  $\mathbf{k}$  dependency, the only relevant parameter is the bare energy  $\epsilon_{\mathbf{k}}$ . In the figure, it is realized that  $P(\omega)$  gives the additional asymmetric broadening. As shown in the last section, the broadening should depend on the dielectric constants such as  $\epsilon_{\parallel}$  and  $\epsilon_{\perp}$ . The resulting curves with the broadening added look comparable to the experimental curves in Liu *et al.*<sup>13</sup> We importantly note that, for experimentally acceptable values of  $\epsilon_{\parallel}$  and  $\epsilon_{\perp}$ , the suitable asymmetric broadening to explain Liu *et al.*'s puzzle is obtained even if the theory is in principle qualitative. Finally, we remark that  $J_{\mathbf{k}}(\omega)$  (not shown here) from off-acoustic  $P(\omega)$  show less broadening up to  $\approx$

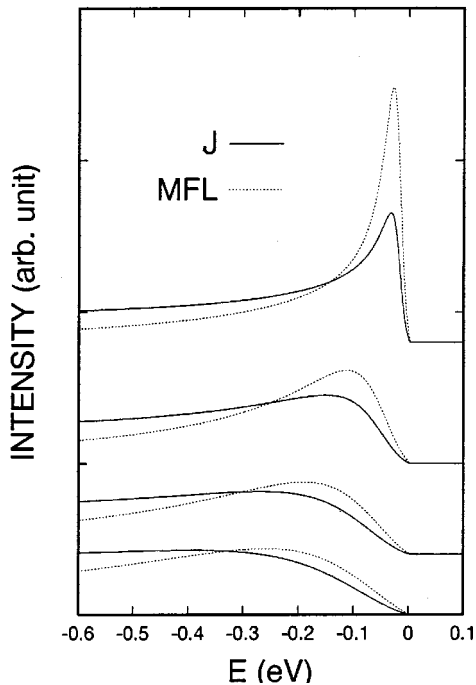


FIG. 6. Final photoemission spectra  $J_{\mathbf{k}}(\omega)$  given for several values of  $\epsilon_{\mathbf{k}} = -0.1, -0.3, -0.5,$  and  $-0.7$  eV from the top curve. “MFL” means  $J_{\mathbf{k}}^{2D}(\omega)$  [Eqs. (17) or (18)] and “J” means the final spectra from Eq. (1). For  $J_{\mathbf{k}}^{2D}(\omega)$ , we take  $g' = 1.0$  and  $\omega_c = 1.0$  eV. All the curves are normalized to have a unit area up to  $-0.6$  eV.

$-0.6$  eV from the quasiparticle peak and also unphysical humps near  $\approx -0.6$  eV compared to the on-acoustic (full) calculation.

## V. CONCLUDING REMARKS

We have investigated photoemission in the system of stacked interacting layers. In the strongly correlated quasi-two-dimensional systems, most of the studies have been concentrated on the physics occurring in the purely two-dimensional layer. Accounting for the three-dimensionality of an actual layered material, we can have a new class of excitation, i.e., the gapless acoustic plasmon. This excitation is quite interesting in the sense that its gaplessness has direct effects on the quasiparticle structure in the photoemission spectra. Particularly, in the high- $T_C$  compounds, theoretical spectral functions, experimental photoemission spectra and their quasiparticle structures have been still far from the overall consensus and being truly controversial.

In this study, combining the general dielectric response formalism of interacting layers and the RPA for a single

layer, we could artificially turn on or off acoustic plasmons and then examine their roles and effects by comparing the on- and off-acoustic calculations. Acoustic plasmons are found to weaken the quasiparticle peak weights and to give asymmetric skew broadening. It is also found that as the interlayer coupling increases ( $\epsilon_{\perp}$  decreases) the acoustic plasmons are more strongly activated in the photoemission process.

The present layered system is thought to be a prototype of the high- $T_C$  superconducting materials. Using the MFL theory for the spectra from the pure two-dimensional layer, we can get the photoemission spectra by convoluting with  $P(\omega)$ . The resulting spectra is more asymmetrically broadened such that it gets comparable to the experimental observation. It is quite fascinating to obtain the suitable asymmetric broadening using typically acceptable values of  $\epsilon_{\parallel}$  and  $\epsilon_{\perp}$ . The degree of broadening is determined by  $\epsilon_{\parallel}$  and  $\epsilon_{\perp}$ . These additionally broadened spectra through  $P(\omega)$  mean that the anomalously large inelastic backgrounds needed to fit the two-dimensional self-energy theory with the experimental data may be ascribed to real intrinsic spectral weights by acoustic plasmon losses to a large extent.

Finally, it would be worthwhile to make a comment on the scope and the outlook of the present model, especially its application to high- $T_C$  cuprates. The RPA-type loss function (corresponding to the model) has never been directly measured yet in the high- $T_C$  cuprates, which means that, as mentioned before, no direct evidence of acoustic plasmons has been available up to now. This may make the validity of the RPA or effects of acoustic plasmons in the system more or less shaky. However, we think that this does not imply necessarily the irrelevance of acoustic plasmons (or the RPA) in the cuprates. One may argue that acoustic plasmons should be expected from quite general arguments, irrespective of the nature of charge carriers, as long as the material can be viewed as a layered conductor and a long range potential is available, i.e., collective charge excitations with  $q \rightarrow 0$  can be realized without energy cost due to a cancellation of electric fields from layers. Further, the model would be still promising since new possibilities to observe acoustic plasmons are discussed.<sup>23</sup>

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<sup>1</sup>For a general review, see Z.-X. Shen and D.S. Dessau, Phys. Rep. **253**, 1 (1995), and references therein.

<sup>2</sup>C.M. Varma, P.B. Littlewood, S. Schmitt-Rink, E. Abrahams, and A.E. Ruckenstein, Phys. Rev. Lett. **63**, 1996 (1989).

<sup>3</sup>P.W. Anderson, Phys. Rev. Lett. **64**, 1839 (1990).

<sup>4</sup>A.L. Fetter, Ann. Phys. (N.Y.) **88**, 1 (1974).

<sup>5</sup>N. Tomita and K. Nasu, Phys. Rev. B **60**, 8602 (1999).

<sup>6</sup>H. Yasuoka, T. Imai, and T. Shimizu, in *Strong Correlation and Superconductivity*, edited by H. Fukuyama, S. Maekawa, and A.P. Malozemoff (Springer-Verlag, Berlin, 1989).

- <sup>7</sup>O. Anderson, R. Manzke, and M. Skibowski, *Phys. Rev. Lett.* **55**, 2188 (1985); R. Claessen, R.O. Anderson, G.-H. Gweon, J.W. Allen, W.P. Ellis, C. Janowitz, C.G. Olson, Z.-X. Shen, V. Eyert, M. Skibowski, K. Friemelt, E. Bucher, and S. Hüfner, *Phys. Rev. B* **54**, 2453 (1996); In the papers, it is shown that no additional backgrounds are observed unlike oxides. It may be understood by noting that atomic polarizabilities of Se, S, and Te are much larger than O. This may imply that the background dielectric constants of the selenides, sulfides, and tellurides are quite large compared to oxides, which probably suppresses the acoustic plasmon effects (see the text).
- <sup>8</sup>L. Hedin and J.D. Lee, *Phys. Rev. B* **64**, 115109 (2001).
- <sup>9</sup>N. Nücker, H. Romberg, S. Nakai, B. Scheerer, J. Fink, Y.F. Yan, and Z.X. Zhao, *Phys. Rev. B* **39**, 12 379 (1989).
- <sup>10</sup>P. Prelovšek and P. Horsch, *Phys. Rev. B* **60**, R3735 (1999).
- <sup>11</sup>S. Doniach and M. Šunjić, *J. Phys. C* **3**, 285 (1970).
- <sup>12</sup>E. Dagotto, R. Joint, A. Moreo, S. Bacci, and E. Gagliano, *Phys. Rev. B* **41**, 9049 (1990); J. Voit, *ibid.* **47**, 6740 (1993); A. George, G. Kotliar, W. Krauth, and M.J. Rozenberg, *Rev. Mod. Phys.* **68**, 13 (1996).
- <sup>13</sup>L.Z. Liu, R.O. Anderson, and J.W. Allen, *J. Phys. Chem. Solids* **52**, 1473 (1991).
- <sup>14</sup>More rigorously,  $P(\omega)$  describes the contribution from extended excitations rather than necessarily from three dimensional excitations. The dominant excitations in strongly correlated two-dimensional systems should be local due to the narrow band feature. In such a sense,  $P(\omega)$  may be said to be from the three-dimensionality.
- <sup>15</sup>L. Hedin, J. Michiels, and J. Inglesfield, *Phys. Rev. B* **58**, 15 565 (1998); for a more extended review, see C.O. Almbladh and L. Hedin, in *Handbook on the Synchrotron Radiation*, edited by E.E. Koch (North Holland, Amsterdam, 1983).
- <sup>16</sup>F. Aryasetiawan, L. Hedin, and K. Karlsson, *Phys. Rev. Lett.* **77**, 2268 (1996); J. Osterwalder, T. Greber, S. Hüfner, and L. Schlapbach, *ibid.* **64**, 2683 (1990).
- <sup>17</sup>L. Hedin, *Phys. Scr.* **21**, 477 (1980); Another detailed analysis would be found in T. Portengen, Ph.D thesis, University of California, San Diego, 1995. It is argued in the thesis that, in a case of the recoiling valence hole in the two-dimensional Fermi sea, the singularity of spectra still remains, but reduces.
- <sup>18</sup>H. Höchster, P. Steiner, G. Reiter, and S. Hüfner, *Z. Phys. B: Condens. Matter* **42**, 199 (1981).
- <sup>19</sup>F. Stern, *Phys. Rev. Lett.* **18**, 546 (1967).
- <sup>20</sup>For the small  $q$  limit, i.e.,  $q, q' \ll 2\pi/c$ , the (inverse) dielectric function should be diagonal.
- <sup>21</sup>D.R. Harshman and A.P. Mills, Jr., *Phys. Rev. B* **45**, 10 684 (1992).
- <sup>22</sup>K. Tamasaku, Y. Nakamura, and S. Uchida, *Phys. Rev. Lett.* **69**, 1455 (1992); S. Tajima, G.D. Gu, S. Miyamoto, A. Odagawa, and K. Koshizuka, *Phys. Rev. B* **48**, 16 164 (1993).
- <sup>23</sup>K. Schulte, D. Dulic, and G.A. Sawatzky, *Physica C* **317-318**, 554 (1999).
- <sup>24</sup>D.B. Tanner and T. Timusk, in *Physical Properties of High-temperature Superconductors*, edited by D.M. Ginsberg (World Scientific, Singapore, 1992).